XXI International Baldin Seminar on High Energy Physics Problems – "Relativistic Nuclear Physics & Quantum Chromodynamics"; Russia, Dubna, September 8 – 15, 2012.

ANALYSIS OF POTENTIAL ADVANCED THORIUM BASED FUEL FOR EPR REACTOR

Marcin Szuta, Andrzej Wojciechowski

National Center for Nuclear research, Otwock-Swierk 05-400, Poland

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

Outline

1. Introduction

- 2. **Preliminary analysis of thorium based fuel application in the EPR reactor.**
- 3. **Conclusions and remarks**

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

1. Introduction

- Analysis of possible ways of reduction of radioactive wastes by transmutation of radioactive long-lived fission products such as ^{99}Tc, ^{129}I and ^{135}Cs and by burning up of transuranic nuclides implies that the reactor core should consist of three zones with fast, epithermal and thermal neutron spectra.
- High flux thermal neutron environment ($\geq 10^{14}$ n/cm^3·s) is expected as the best way for the transmutation of most of the radioactive waste to stable or short-lived nuclides and for increasing the probability for fission such actinides as ^{237}Np and ^{238}Np.
- Since we are not able to construct such a reactor core, we focus our attention on the thorium-uranium fuel cycle as a prophylactic way of energy production where the actinides radio-toxicity of the wastes is about three orders of magnitude smaller than in the case of classical PWR [1] reactors .

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

1. Introduction

- For the thorium-uranium fuel cycle the actinides radio-toxicity of the wastes is about three orders of magnitude smaller than in the case of classical PWR [1] reactors.
- **This is clearly demonstrated on Fig. 1. beside (**Radio-toxicity for various cycles [1].)
- Comparison of the potential radio-toxicity of fission products in **Fig. 2 beside** [2] and in **Fig.1** after 250 years where it can be neglected with the actinides radio-toxicity for the thorium-uranium fuel cycle after 10 years let us infer that they are nearly equal to the uranium ore.

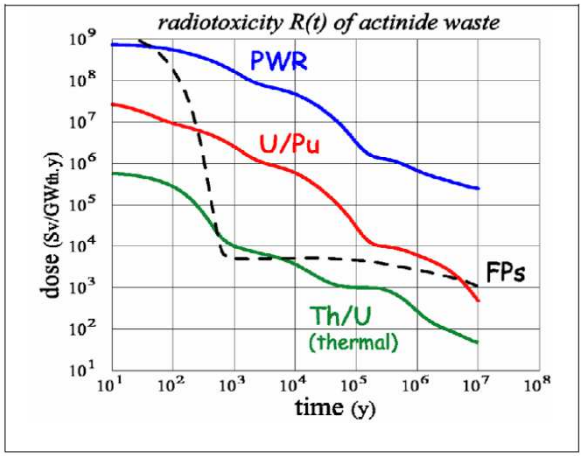


Fig. 1 [1]

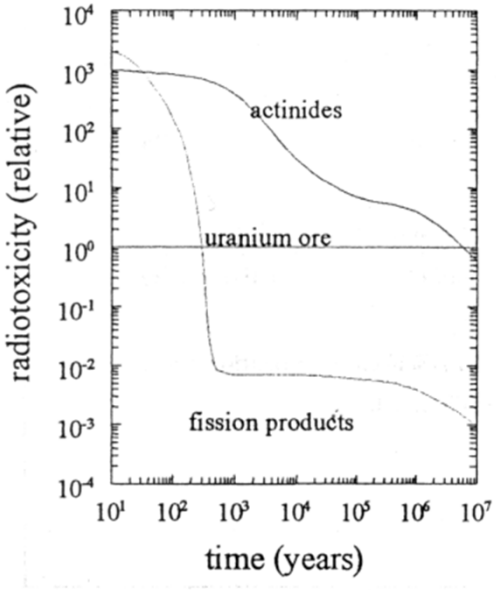
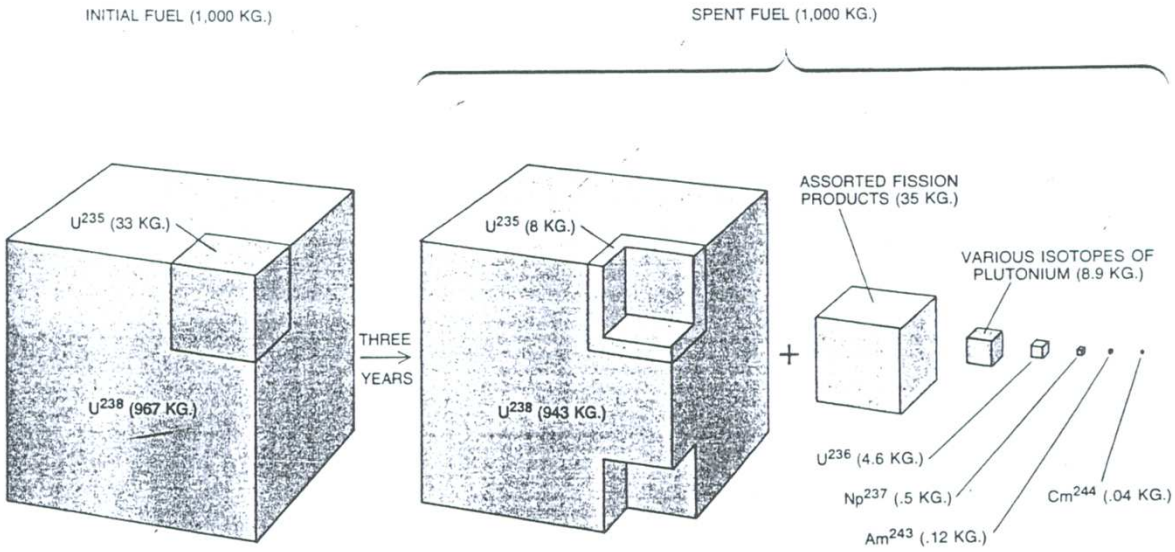


Fig. 2 [2]

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

1. Introduction



- Fig. 3. Balance of fission products and actinides in one tone of uranium fuel enriched to 3.3 % ^{235}U after three years of exploitation in the energetic reactor [3]. Burn-up of about 40 GWD/tU.

- Assessment of radioactive waste generated in the energetic reactors.
- The energetic reactor of 1000 MW_e electric nominal power generates about 10 tons per year of the spent fuel. Using the data given in Fig. 3 [3] the quantitative isotopic composition of this fuel is as follows: 9430 kg U-238, 80 kg U-235, 350 kg of fission products, 89 kg of different isotopes of plutonium and 46 kg U-236, 5 kg Np-237, 1.2 kg of Am-243 and 0.4 kg Cm-244. In the case of Th-U cycle, the amount of actinide waste is three order less.

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

1. Introduction

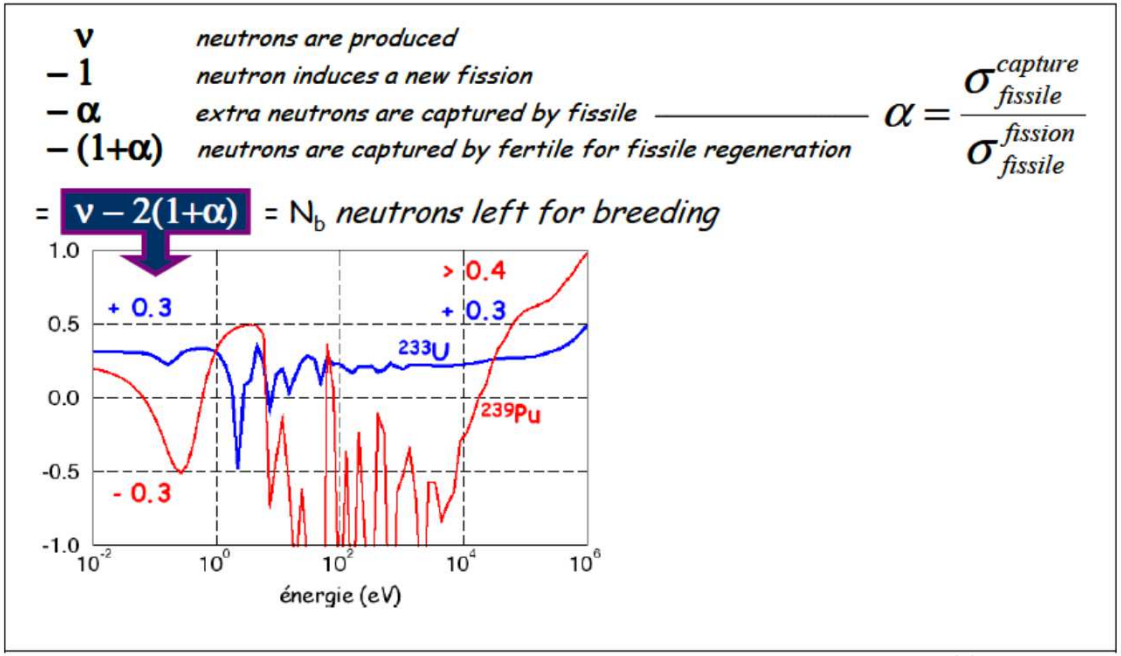


Fig. 4. Available neutrons for breeding both for ^{233}U and ^{239}Pu [4].

The^{232}Th – ^{233}U fuel cycle let us obtain breeding of fissile atoms both in fast, epithermal and thermal neutron spectra (see Fig. 4) [4]. The authors [4] underline that as long as the available neutron numbers for breeding(N_b) is always slightly larger than 0, breeding is possible. This is the case for the whole neutron energy spectrum for uranium 233 where N_b is always equal 0.3.

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

1. Introduction

- However the number of neutrons available for breeding (Nb) presented in paper [1] is equal to about 0.1 **(see Fig. 5 beside).** In Fig. 5 the breeding feasibility for different fuel cycles is considered.
- While the uranium 238-plutonium 239 fuel cycle requires fast neutrons to be sustainable, the thorium 232-uranium 233 fuel cycle is sustainable with either thermal neutrons or fast neutrons.

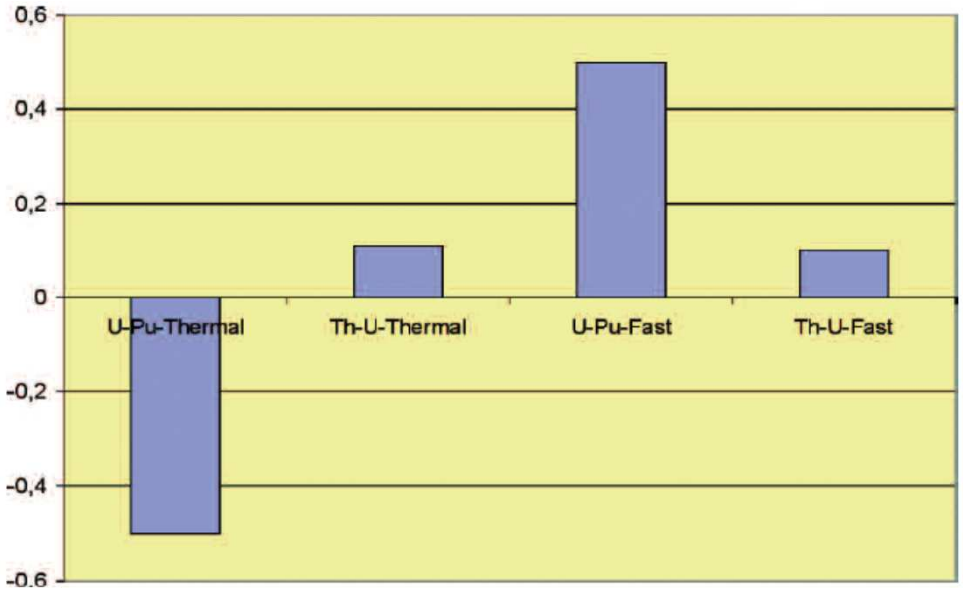


Fig. 5. Number of neutrons available for breeding (Nb) in the uranium-plutonium and the thorium-uranium cycles with thermal and fast neutron spectra. Breeding is impossible for negative values of (Nb) [1].

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

1. Introduction

- As thorium based fuels have benefits in terms of radio-toxicity there are however challenges in terms of reprocessing the spent thorium based fuel. The database and experience of thorium fuels and thorium fuel cycles are very limited , as compared to UO_2 and $(U,Pu)O_2$ fuels, and need to be augmented before large investments are made for commercial utilization of thorium fuels and fuel cycles [5].
- That is why, as the first step of studying the thorium application feasibility, once through thorium based fuel cycle analysis for energy production and radioactive waste transmutation was undertaken, which does not require the technically difficult reprocessing of the spent fuel.
- The idea is to perform the thorium based fuel application in the open cycle feasibility study in the existing light water reactors with minimum modifications in order to exploit them.

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor.

- In many institutes the possibility of using thorium [6-13] in reactors was investigated. Some calculation results of thorium based fuel application in the EPR reactor are presented in our work [13].
- The main aim of this work is to show dependence of thorium burn-up in different places of reactor core. It is needed to determine optimal conditions of U-333 productions (position in reactor core and burning period of time). To investigate this problem we used European Pressurized Reactor EPR[13-15].

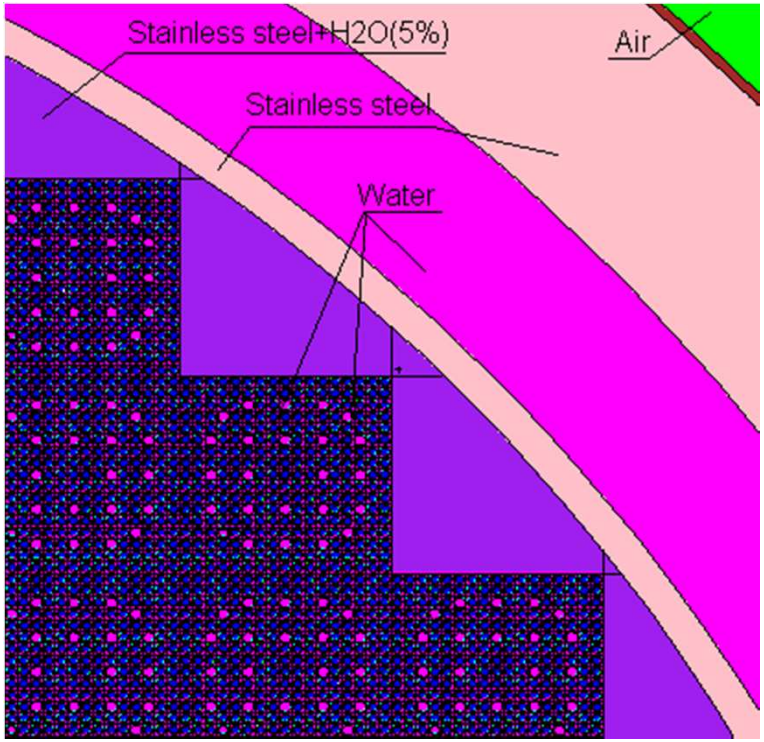


Fig. 6. Vertical cross section of EPR core fragment presenting several UO_2 fuel assemblies neighboring the reflector (stainless steel +H_2O(5%)).

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor.

- Applying the Monte Carlo methodology (MCNPX 2,6) we have calculated the neutron multiplication factor k_{eff} in function of burn-up for the reactor core loaded with 241 uranium fuel assemblies (maximal amount) as in Fig. 6 exploited in constant power of 5000 MW_{th}. Burn-up in the abscissa axis denotes the average burn-up. This calculation presented in Fig. 7 is performed in order to find a reference curve in further simulations.

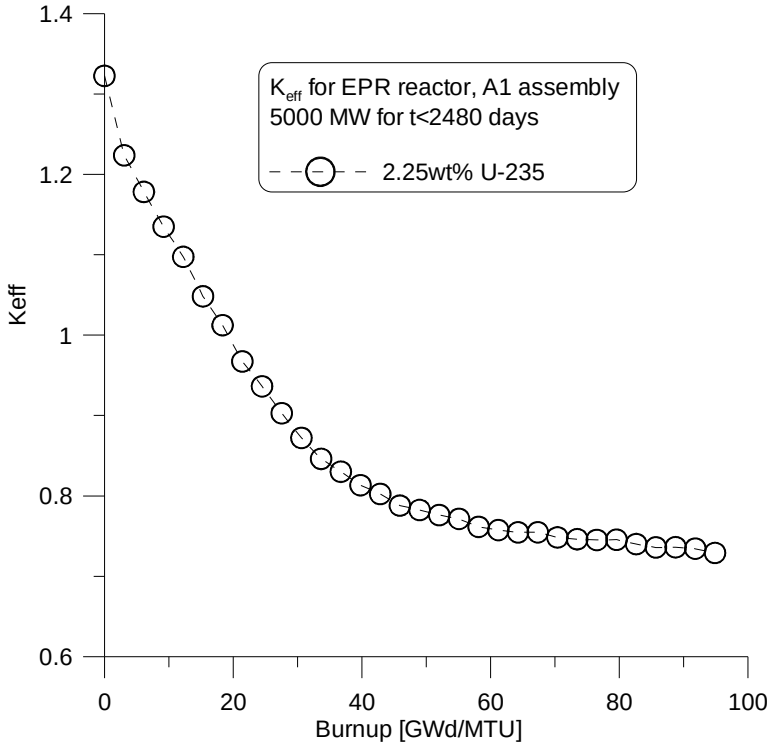


Fig. 7. Neutron multiplication factor versus burn-up for the EPR reactor core as in Fig. 6.

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor.

- We have expected that a peripheral part of the reactor core is the best location of thorium dioxide rods to convert the fertile thorium ^{232}Th into fissile uranium ^{233}U.
- That is why we performed computer simulation for the configuration (189 uranium and 52 thorium assemblies) presented in **Fig. 8 beside** and calculated the neutron multiplication factor k_{eff} in function of irradiation time for constant power of 5000 MW_{th} and compared it with the k_{eff} for solely 189 uranium fuel assemblies.

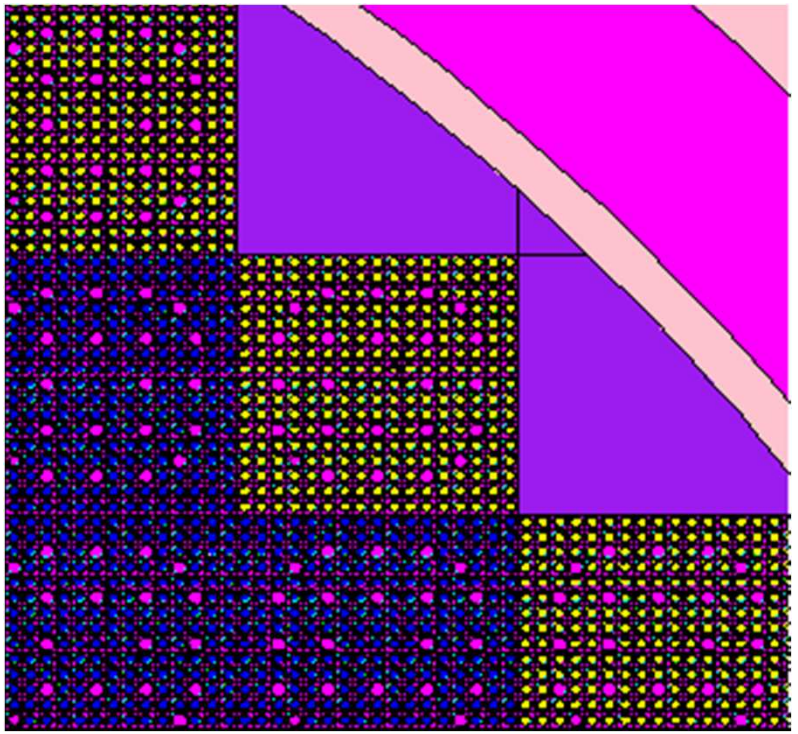


Fig. 8. Location of fuel assemblies in the EPR core: peripheral assemblies (52) neighboring the reflector – ThO_2, remaining assemblies (189) – UO_2.

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor

- Variation of the k_{eff} in function of time (burn-up) for the two core versions are presented in Fig. 9 beside.
- It is clearly seen that impact of the peripheral thorium fuel element assemblies on the neutron multiplication factor is insignificantly small for the zero burn-up.
- Comparing Fig. 7 and Fig. 9 we can see also that impact of the uranium fuel element assembly on the initial k_{eff} is unimportant (small) either, because the difference is equal to about 0.03 what is consumed during several days of reactor work on 5 GW_{th} power.

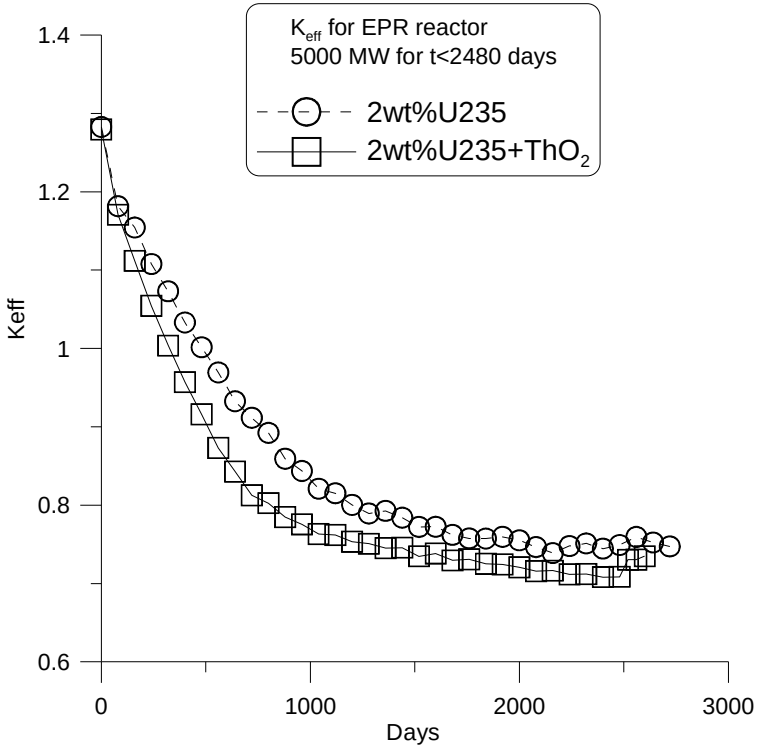


Fig. 9. Variation of k_{eff} in function of irradiation time for the core configurations presented in Fig. 8 and the configuration when solely the uranium assemblies (189) are exploited on thermal power 5000 MW_{th}.

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor

- Assuming that all the 241 thorium assemblies with 1.03 wt% concentration of uranium 233U were loaded into EPR reactor core, we performed computer simulation of neutron multiplication factor keff in function of burn-up for the 5 GWth power and compared it with the same simulation but for the uranium 233U concentration equal to 1.48 wt% (**see Fig. 10** beside).

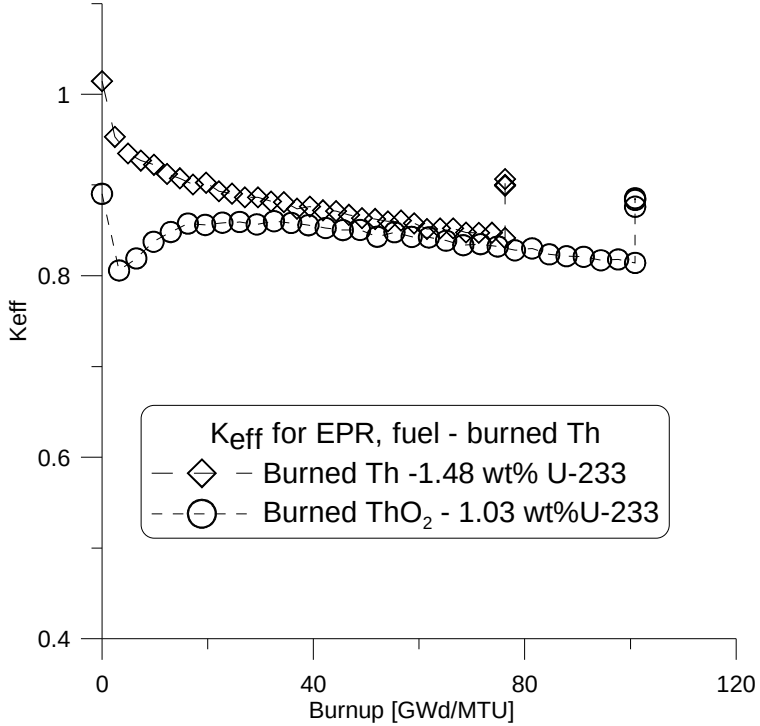


Fig. 10. Neutron multiplication factor keff in function of burn-up for thorium fuel assemblies with 1.03 wt% and for 1.48 wt% 233U concentration of EPR reactor core exploited on the 5 GWth power

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor

- We expect that the initial uranium 233U concentration once is above and once is below the saturation concentration what explains the different shapes of these two curves. In the beginning of the burning process the U-233 concentration sharply decreases in both cases because the U-233 is not being produced due to the fact that the Pa-233 just started to appear.
- The half-life of Pa-233 totals about 27 days. Therefore the production of U-233 will appear with a delay.
- In case of a higher initial uranium 233U concentration level (1.48 wt%), the U-233 concentration tends monotonically to a saturation value.
- Whereas, in case of a lower initial uranium 233U concentration level (1.03 wt%), the U-233 concentration at the beginning decreases and then increases again to the saturation value. The saturation value of 233U concentration will be considered later.

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor

- In order to clarify the different shapes of neutron multiplication factor keff presented in Fig. 10 and to investigate the uranium233U concentration dependence on fission rate and time of irradiation we assumed that the EPR fuel rod is filled with UO2 and ThO2 forming in the reactor core several fuel layers alternately located.
- Symmetrically from the fuel rod center, five thorium dioxide layers 20 cm thick are located (distributed) in the following ranges 30-50cm, 70-90cm, 110-130cm, 150-170cm, 190-210cm. The remaining ranges are filled with enriched 2.25 wt% uranium dioxide (**see fig.11 beside**).
- This kind of fuel rod arrangement is technically feasible**.**

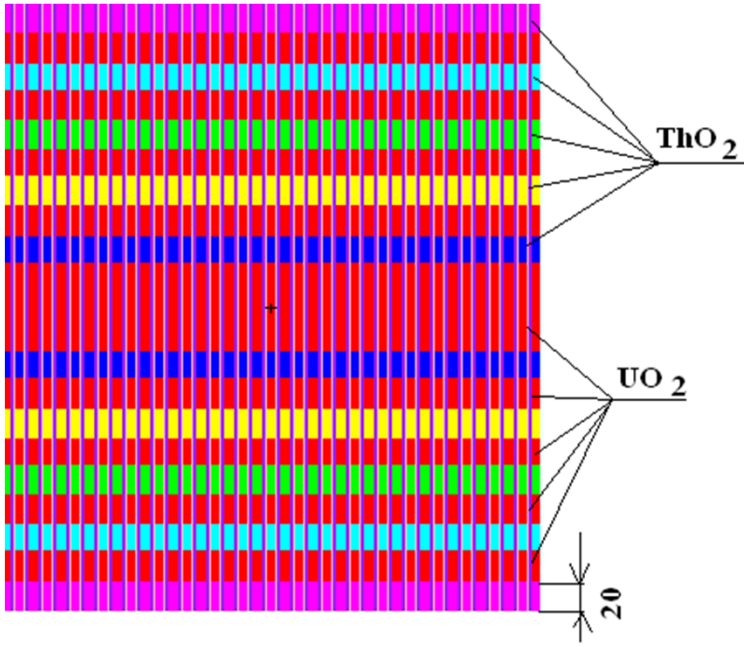


Fig. 11. Longitudinal cross section of EPR core. Different size fuel layers in reactor core. Color red means UO2; Colors: blue, yellow, green, white blue, pink means ThO2

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor

- "Ab initio" calculations compared with the experimental data show that the lattice constants in the Th1–*x*U*x*O2 compound decrease linearly with increasing mole ratio *x* what means that the uranium dioxide lattice constants are smaller than the thorium dioxide lattice constants (**see Fig. 12** beside) .
- This leads to the conclusion that the uranium dioxide and thorium dioxide-based fuel pellets can be in different proportions and alternately located in the fuel rod because U-233 increase in the thorium pellet causes a decrease its volume what, in turn do not generate stresses between the rod clad [16].

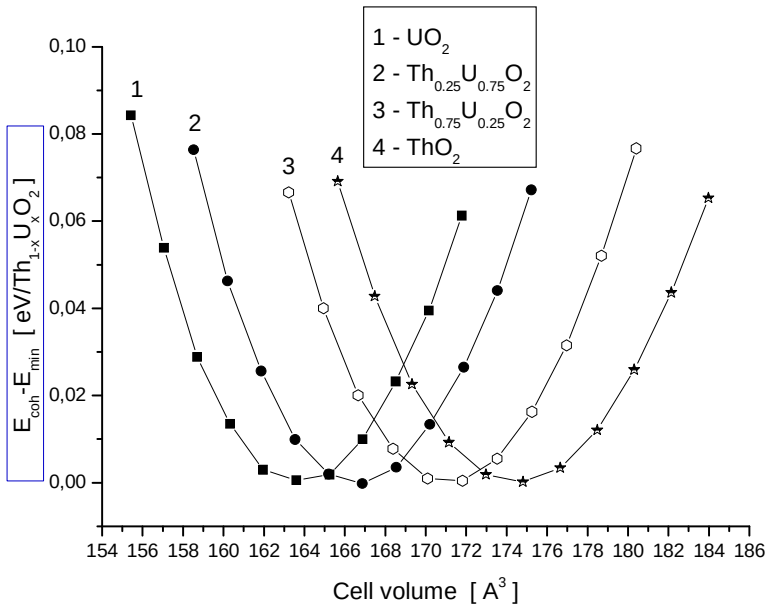


Fig. 12. Variation of the cohesive energy with cell volume for Th1–*x*U*x*O2 [16]

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor

- Fig. 13 shows comparison of keff for reactor core loaded with UO2 (2.25%U-235) solely and with ThO2 + UO2 as described above (see fig. 11). This comparison shows that using the ThO2 + UO2 fuel, the keff decreases about 0.1 from 1.31 to 1.21 at the beginning of exploitation.
- Multiplication factors keff for both reactor configurations decrease similarly fast during burning at the beginning and stabilizes on the value about equal 0.8 after 1000 days of reactor operation (5000 MWth). While the keff for the core loaded solely with UO2 (2.25%U-235) still further slightly decreases , the multiplication factor for the configuration with thorium (Fig.11) does not decrease (self sustaining system).

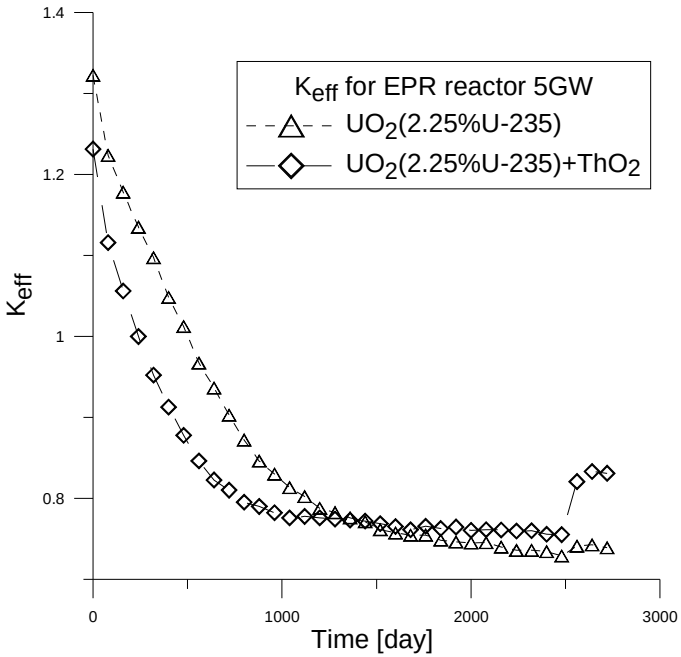


Fig. 13. Comparison of keff for EPR reactor core loaded with UO2 (2.25%U-235) solely and with ThO2 + UO2 as described in Fig. 11

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor

- The concentration of U-233 tends to the saturation value with reactor operation time and sharply increases after cutting off the power (see Figs. 14). In this reactor the saturation concentration of U-233 amounts to about 0.0129 during its work (Fig. 14).
- During the burning time equal 2500 days only three thorium layers (30-50cm, 70-90cm and 110-130cm) achieved saturation concentration.
- The saturation value of U-233 concentration practically does not depend on the power density (Fig. 14) ,while the kinetics of reaching the saturation value strongly depends on the power density (neutron flux). The higher is the neutron flux the more rapid increase of U-233 concentration is observed.

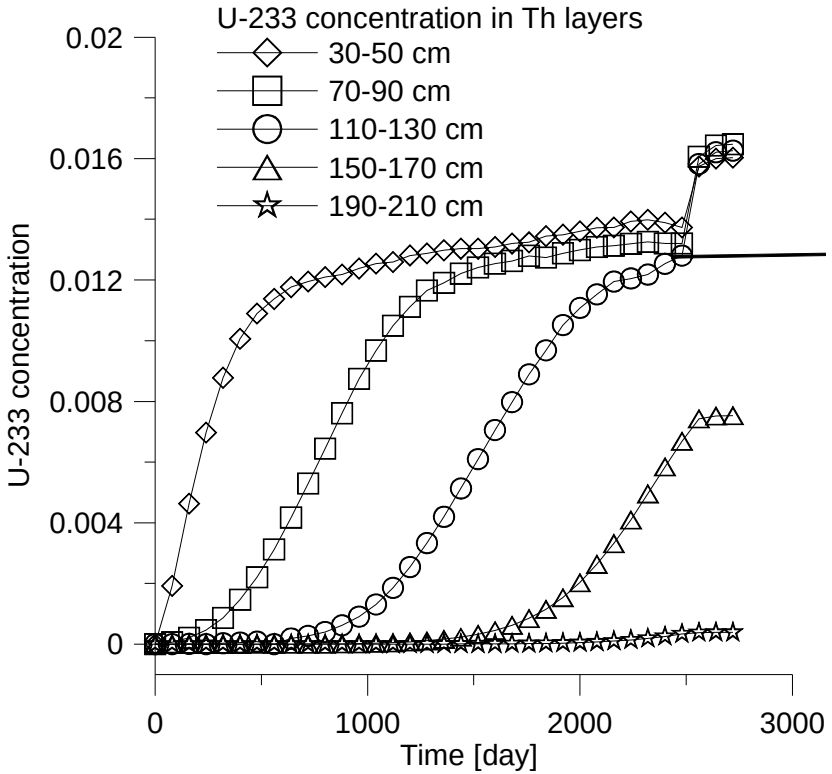


Fig. 14. The uranium233U concentration versus reactor operation time for five thorium dioxide layers 20 cm thick as in Fig. 11

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor

- The five fertile thorium fuel layers alternately located with the uranium dioxide in the fuel rod (see Fig. 11) are irradiated with different neutron flux. The closer is the fertile thorium fuel layer to the center of the reactor core the higher is the neutron flux. The final concentration of U-233 (Fig. 14) consists of two parts: a ‘‘power on’’ concentration and a ‘‘power off ‘‘one. The total mass (and concentration) of the ‘power off’ U-233 is equal to the total mass (and concentration) of Pa-233 at the moment of cutting off the power supply.
- In general, we can infer that the 233U concentration tends to saturation value which does not depend on power density while the kinetics of reaching the saturation value depends on it.

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor

- Fig. 15 shows that the concentration of Pa-233 strongly depends on the neutron flux density. The higher is the neutron flux density the more rapid increase of Pa-233 concentration is observed (occurs) and the higher maximum protactinium concentration is reached in function of reactor operation. In this case the protactinium concentration does not reach saturation. Please note, that the fixed 5 GWth power of the reactor (in U – Th fuel rods) does not mean the fixed neutron flux density in the U – Th rods. The neutron flux density in the rods is relatively complicated function of time and burn-up of thorium.

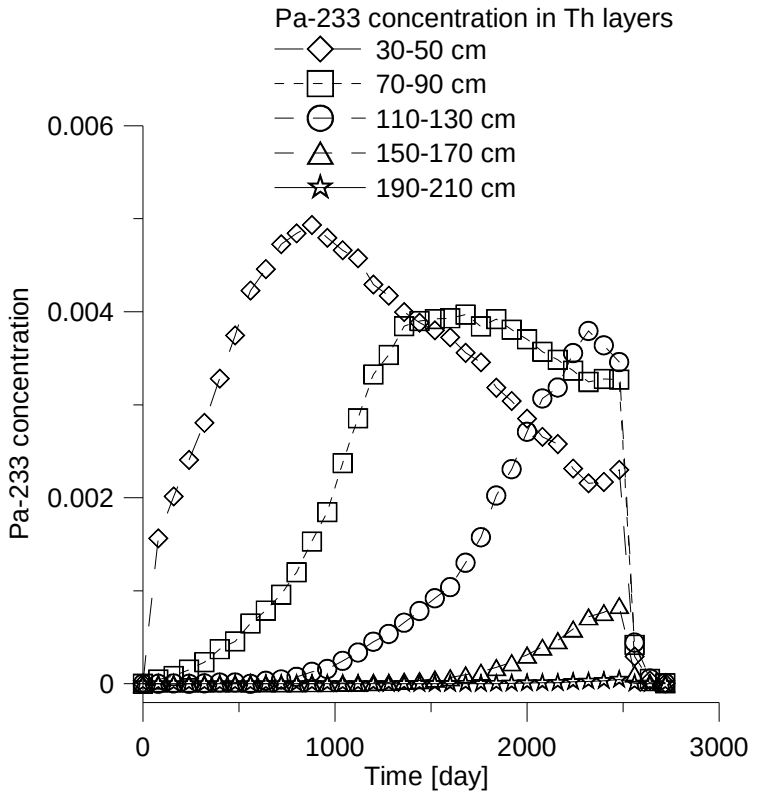


Fig. 15. Concentration of Pa-233 versus reactor operation time (5 GWth) for five thorium dioxide layers 20 cm thick as in Fig. 11.

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

2. Preliminary analysis of thorium based fuel application in the EPR reactor

- Initially, the thorium dioxide layers only absorb the neutrons produced in the uranium dioxide layers and after with time of irradiation the thorium layers produce themselves the neutrons due to conversion of fertile thorium into fissile uranium and take on the power load (see Fig. 16) what in turn decreases the neutron flux density. The growing smaller amount of thorium nuclei in function of burn-up influences also on the protactinium concentration. This explains why the protactinium concentration does not tend to the saturation value.
- In this work the concentration of Pa-233 or U-233 is defined as a relation of the mass of Pa-233 or U-233 to mass of all actinides in unit of volume.

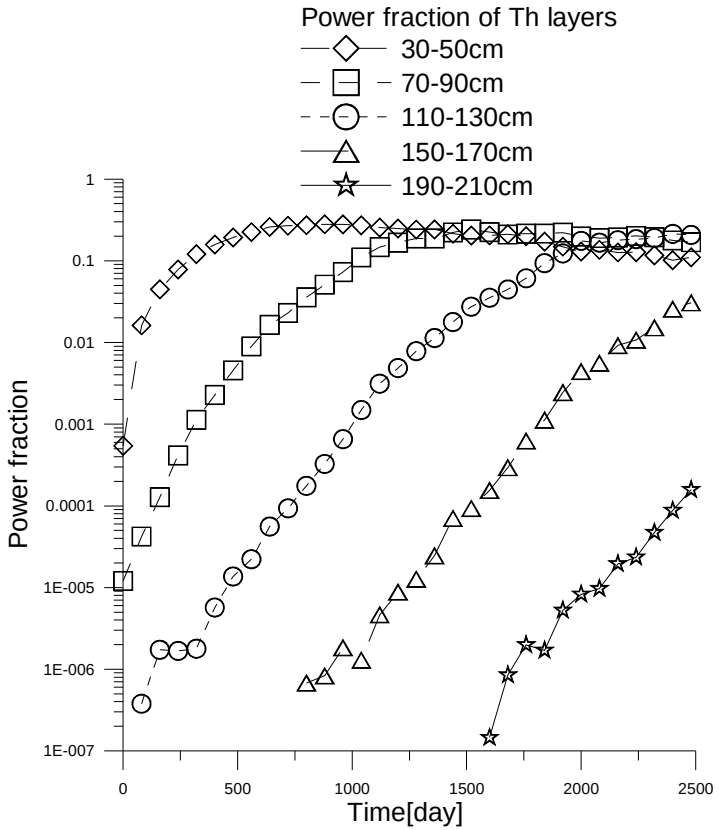


Fig. 16. Power fraction versus reactor operation time (5 GWth) for five thorium dioxide layers 20 cm thick as in Fig. 11

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

3. Conclusions and remarks

- Conversion of the fertile thorium 232Th into fissile uranium 233U in the peripheral part of the EPR reactor core is ineffective since the neutron flux is comparatively small.
- For initial concentration of U-233 equal to saturation value (1.29 wt%) the EPR reactor does not reach criticality, scarcely for 1.5 wt% the reactor becomes slightly overcritical what permits to exploit the reactor during certain time. This concentration value is a sum of the saturation concentration with the “power off” concentration**.** It means that once through thorium fuel cycle can be reached with difficulty.
- The saturation value of U-233 concentration practically does not depend on the power density. While the kinetics of reaching the saturation value strongly depends on the power density (neutron flux).
- Whereas the available neutron numbers for breeding in the thorium-uranium fuel cycle is only a little above zero (0.1 – 0.3, the value is not given precisely in the open literature) we can infer that in the accelerator driven system (ADS) the effect of breeding can be easier utilized in a more flexible way for higher burn-up of the fuel giving impact on economy improvement.
- Applying the thorium-uranium fuel cycle the radio-toxicity of the wastes is about three orders of magnitude smaller than in the case of classical PWR reactors.

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

4. References

- [1] Sylvain David, Elisabeth Huffer and Herve Nifenecker; Revisiting the thorium-uranium nuclear fuel cycle; Europhysics News , Volume 38, number 2 (2007) 24 – 27; Article available athttp://www.europhysicsnews.org or http//dx.doi.org/10.1051/EPN:20072007.
- [2] D. Haas: *The fuel development programme ADS at ITU*; Topical Day on "R&D activity in support of ADS development held at SCK·CEN – Mol on October 24th 2000.
- [3] B.L. Cohen, The Disposal of Radioactive Wastes from Fission Reactors, Scientific American, June 1977 vol. 236, nr 6
- [4] Christian Le Brun, Ludovic Mathieu, Daniel Heuer and Alexis Nuttin; Impact of the MSBR concept technology on long-lived radio-toxicant and proliferation resistance; LPSC 05-81,document IAEA
- [5] IAEA; Thorium fuel cycle – Potential benefits and challenges; IAEA-TECDOC-1450, IAEA Vienna May 2005.
- [6] Yu Jiyang, Wang Kan, You Songbo, Jia Baoshan, Shen Shifei, Shi Gong, et al., Thorium Fuel Cycle of a Thorium-based Advanced Nuclear Energy System Progress in Nuclear Energy, Vol. 45, No. 1, pp. 71 - 84, 2004.
- [7] Permana Sidik, Naoyuki Takaki, Hiroshi Sekimoto, Power density effect on feasibility of water cooled thorium breeder reactor, Progress in Nuclear Energy 50 (2008) 308-313.
- [8] A. Lafuente, M. Piera, Exploring new coolants for nuclear breeder reactors, Annals of Nuclear Energy, 37 (2010), 835–844

Analysis of Potential Advanced Thorium Based Fuel for EPR Reactor.

4. References

- [9] Chang Joon Jeong *, Chang je Park, Won Il Ko, Dynamic Analysis of a thorium fuel cycle in CANDU reactors, Annals of Nuclear Energy 35 (2008) 1842–1848
- [10] M. Matsunaka , S. Shido, K. Kondo, H. Miyamaru, I. Murata; Burnup calculation of fusion–fission hybrid energy system with thorium cycle Division of Electrical, Electronic and Information Engineering, Graduate School of Engineering, Osaka University, Yamada-oka 2-1, Suita, Osaka 565-0871, JapanReceived 1 August 2006; received in revised form 22 March 2007; accepted 22 March 2007. Available online 7 May 2007
- [11] J. Breza, P.Darˇílek , V. Necˇas Study of thorium advanced fuel cycle utilization in light water reactor VVER-440 Annals of Nuclear Energy, Annals of Nuclear Energy 37 (2010) 685–690
- [12] S. Mittag, S. Kliem, Burning plutonium and minimizing radioactive waste in existing PWRs, Annals of Nuclear Energy, Volume 38, Issue 1, January 2011, Pages 98-102
- [13] M. Szuta, A. Wojciechowski; Przyczynek do analizy wykorzystania paliwa na bazie toru w zestawach podkrytycznych sterowanych akceleratorem i w reaktorze EPR, (Contribution to the analysis of thorium based fuel application in the accelerator driven systems and in the EPR reactor).Science Conference, Mądralin near Warsaw, Poland, 13 – 14 June 2011
- [14] S.Kilim, Basic Data of the EPR Reactor, Raport IEA, B-16, 2009
- [15]http://adamswebsearch2.nrc.gov/idmws/ViewDocByAccession.asp?AccessionNumber=ML092450764 AREVA Design Control Document Rev. 1 - Tier 2 Chapter 04 - Reactor - Section 4.3 Nuclear Design, ML091671461 2009-05-29
- [16] L. Dąbrowski, M. Szuta; Local structure and cohesive properties of mixed thorium dioxides calculated by “ab initio” method; Nukleonika 2012, 57(1), 101-107.

- Thank you for the attention.